

Docket No.: 6920/1029-US0  
(PATENT)

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

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In re Patent Application of:  
Hirofumi Ito et al.

Application No.: 10/595,622

Confirmation No.: 3648

Filed: June 8, 2006

Art Unit: 1793

For: CATALYST, PROCESS FOR PREPARING  
THE CATALYST AND PROCESS FOR  
PRODUCING LOWER HYDROCARBON  
USING THE CATALYST

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Examiner: Elizabeth D. Wood

DECLARATION PURSUANT TO 37 CFR §1.132

MS Amendment  
Commissioner for Patents  
P.O. Box 1450  
Alexandria, VA 22313-1450

Sirs:

I, Atsushi Okita, hereby declare as follows:

I am a citizen of Japan and over 21 years of age. I am a graduate of Tokyo Institute of Technology, located on 4529 Nagatsuda, Midori-ku, Yokohama, Kanagawa, Japan and received a degree in Catalyst Chemistry from the Department of Environmental Chemistry and Engineering. I have been employed by JGC Corporation since April 01, 2005 and I have been conducting research in the field of olefin catalysts for five (5) years. I have reviewed the Office Action mailed January 13, 2010 and the references cited therein. It is my understanding that the Examiner has rejected the claims of the application based on his belief that combining the teachings of Japanese Laid-Open Patent Application No. 61-58812 with US Patent No. 4,544,793 to Okado

I have conducted experiments which demonstrate that the features of the present invention differ significantly from the features obtainable by the techniques disclosed by the Japanese Laid-Open Patent Application No. 61-58812 patent in combination with and US

4,544,793 to Okado. In fact, these experiments demonstrate that the catalyst of the present invention cannot be produced from the combination of Japanese Laid-Open Patent Application No. 61-58812 ('812 patent) and US 4,544,793 to Okado.

Attached is Experimentation Report 1 (Exhibit 1) which documents the experiments and shows that the catalyst according to the present invention has a particle size which is smaller than that of the catalyst that results from any obvious combination of the '812 patent and Okado references.

In the January 13, 2010 Office Action, the Examiner stated that it is well known in the art of zeolite synthesis to employ seed crystals to produce final compositions having crystals of consistent and desired particle size. The Examiner cites the '812 patent as teaching that ZSM-5 seed crystals are known to be used during the synthesis of zeolite materials. Furthermore, the Examiner considers that arrival at the optimum or desired ranges to be merely a matter of routine experimentation due to the fact that the compositions of the prior art and the instant claims are similar.

Experimentation Report 1 page 1, ¶¶ 2-4, clearly shows the results of the process as described in the pending claims. The zeolite used was commercially available product, and of which average particle size was 0.5 $\mu$ m. There is a distribution among the crystal sizes, and the crystals do not have the same size. The end product of the process as described in the pending claims of the present invention have has an average particle size of 1.5 $\mu$ m Experiment 1, particularly at, ¶ 2 and Fig 2 clearly show that the process as described by the present invention yields a MFI structure zeolite having an average particle size of 1.5 $\mu$ m.

In the same January 13, 2010 Office Action, the Examiner judged that the present invention is obvious from the combination of US4,544,793 (Okado) and JP 61-058812 under section 103(a). So, we performed an experiment for demonstrating that the advantageous effect of the present invention (i.e. that the average particle size of catalyst is small) which cannot be obtained from an obvious combination of Japanese Patent Reference No. JP59-97523 and Japanese Patent Reference JP61-058812. Japanese Patent Reference No. JP59-97523, which is the Japanese counterpart of US 4,544,793, discloses a process for preparing catalyst. Japanese Patent Reference No. JP59-97523 discloses a seed crystal,

Experimentation Report 1, pg 2, details a zeolite catalyst produced by a process combining the '812 patent and Okado references. The average particle size of the seed crystal A was 0.5 $\mu$ m. It is impossible to control the size of zeolite to be synthesized by optimizing the size of seed crystal, because of the seed crystal is decomposed once in hydrothermal synthesis of zeolite occurs and once zeolite is synthesized. This is due to the fact that the conditions of hydrothermal synthesis affect the size of zeolite to be synthesized, etc.

As shown at pg 2, ¶¶ 4-7, the catalyst particles that result from a combination of the '812 and Okado references have an average particle size of 5.0 $\mu$ m. Fig 3 clearly shows that the particles produced by a combination of the prior art are significantly larger than those produced by the instant claims.

Pending claim 3 of the present application recites a catalyst particle having an average diameter of 0.05 - 2.00 $\mu$ m. The size of a catalyst particle is directly related to its durability and usefulness. Smaller diameter particles are more effective. It is clear from Exhibit 1 that the cited prior art, in obvious combination, as disclosed by the Examiner, fails to teach a catalyst particle diameter having this range. Therefore, it is clear from Exhibit 1 that the prior art combination produces a product that is inferior to that disclosed in claim 3.

A person skilled in the art could not employ mere "routine experimentation" and hope to achieve the range described in amended claim 3. Specifically, in order to achieve the range as described in claim 3, there would be need to be a directed effort to obtain this range. Exhibit 1 clearly shows that combining the prior art references results in an average particle diameter that is 10 times that which is found in the amended claim 3. Without foreknowledge of the effective range resulting from the instant claims, i.e. the specific combination of seed crystal properties and process; any prior art combination must be judged on the actual outcome of a combination. Here, the prior art combination produces a product so outside the claimed range that mere experimentation would fail to close the gap.

Exhibit 1 demonstrates that the process as described in claim 3 of the instant application produces a product having characteristics that are not obvious in light of the combined prior art.

I further declare that all statements made herein of my own knowledge are true, and that all statements made on information and belief are believed to be true; and further that these statements are made with knowledge that willful false statements and the like so made are punishable by fine or imprisonment or both, under § 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the instant application or any patent issued thereupon.

June 4, 2010

Date

Atsushi Okita

Atsushi Okita

## Experimental Report 1

Result of catalyst preparation in the case in which Japanese Patent Laid-Open No. 61-58812 is combined with Japanese Patent Laid-Open No. 59-97523.

### [Experiment 1]

〈 Preparation method of zeolite catalyst disclosed in JGC patent 〉

Preparation was conducted through a preparation method which has been improved based on the preparation example disclosed in the Document No.1(Japanese Patent Publication No. 63-35570 bulletin).

Zeolite raw material liquid consisting of 9.50g of  $\text{Al}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$  and 10.92g of  $\text{Ca}(\text{CH}_3\text{COO})_2 \cdot \text{H}_2\text{O}$  was dissolved in 750g of water, and then to the resultant solution, a solution obtained by dissolving 500 g of Cataloid Si-30 water-glass (produced by Shyokubai-Kasei Kogyo C., Ltd.) in 333g of water, 177.5g of an aqueous solution of NaOH having 6 mass %, 317.6g of an aqueous solution of tetra propyl ammonium bromide having 21.3 mass %, and 15.0g (i.e. it is amount which is equivalent to 10 mass % of the amount of zeolite catalyst which is synthesized without using seed crystal) of an ammonium type MFI structure zeolite having average particle size of  $0.5\mu\text{m}$  (produced by Zeolyst Co., Ltd., of which Si/Al atomic ratio is 70) as a zeolite seed crystal were added, while agitating, to obtain an aqueous gel mixture.

Subsequently, the resultant aqueous gel mixture was put in 3L autoclave container, and was agitated at  $160^\circ\text{C}$  under self-pressure for 18 hours to conduct hydrothermal synthesis.

After a white solid product derived from hydrothermal synthesis was filtered and washed with water, the resultant product was dried at  $120^\circ\text{C}$  for 5 hours, and was burned at  $520^\circ\text{C}$  in the air for 10 hours.

The resultant burnt product was immersed in a 0.6N hydrochloric acid, and was agitated at room temperature for 24 hours to make the type of zeolite be proton type.

Subsequently, the resultant product was filtered and washed with water, and then was dried at  $120^\circ\text{C}$  for 5 hours, then was burned at  $520^\circ\text{C}$  in the air for 10 hours to obtain proton type alkaline earth metal containing MFI structure zeolite catalyst.

[Production method of seed crystal A of Japanese Patent Laid-Open No. 61-58812]

Seed crystal was produced by the production method of seed crystal A disclosed in Japanese Patent Laid-Open No. 61-58812.

#### Seed crystal A

1.1g of sodium hydroxide solid and 9.0g of aqueous sodium aluminate ( $\text{Na}_2\text{O}$ : 26wt%,  $\text{Al}_2\text{O}_3$ : 20wt%,  $\text{H}_2\text{O}$ : 54.3wt%) were added to 150g of pure water to obtain uniform aqueous solution, and then to the resultant aqueous solution, 30g of white carbon (produced by in Tosoh Silica Co., Ltd., brand name of Nip Seal VN-3,  $\text{SiO}_2$ : 87.7wt%,  $\text{Al}_2\text{O}_3$ : 0.5wt%) was added, with stirring. The resultant raw material mixture was allowed to stand in an autoclave held at 160°C for 72 hours to be crystallized, and then the resultant reaction product was subjected to solid-liquid separation, washed with water sufficiently, and was dried at 110°C. Thereafter, the resultant product was crushed in a mortar. X-ray pattern of the resultant product is shown in Fig. 1. Two peaks were observed within a range of 3.70 to 3.86Å.

[Combination of the seed crystal A disclosed in Japanese Patent Laid-Open No. Sho.61-58812 with Japanese Patent Laid-Open No. 59-97523]

Zeolite raw material liquid consisting of 0.60g of  $\text{Al}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$  and 0.73g of  $\text{Ca}(\text{CH}_3\text{COO})_2 \cdot \text{H}_2\text{O}$  was dissolved in 72g of water, and then to the resultant solution, a solution obtained by dissolving 48g of Cataloid Si-30 water-glass (produced by Shyokubai-Kasei Kogyo C., Ltd.) in 32g of water, 16.9g of an aqueous solution of NaOH having 6 mass %, 30.5g of an aqueous solution of tetra propyl ammonium bromide having 21.3 mass %, and 3.0g (i.e. it is amount which is equivalent to 1.5 mass % of the raw material mixture) of the seed crystal A which was synthesized by the same method as that disclosed Japanese Patent Laid-Open No. Sho.61-58812, as a zeolite seed crystal, with agitating, to obtain an aqueous gel mixture.

Subsequently, the resultant aqueous gel mixture was put in 3L autoclave container, and was agitated at 160°C under self-pressure for 18 hours to conduct hydrothermal synthesis.

After a white solid product derived from hydrothermal synthesis was filtered and washed with water, the resultant product was dried at 120°C for 5 hours, and was burned at 520°C in the air for 10 hours.

The average particle size of the prepared catalyst was 5.0μm.

[Explanation of drawings]

[FIG. 1]

It is an X-ray pattern of seed crystal A.

[FIG. 2]

It is electron micrograph of synthesized zeolite in Experiment 1.

[FIG. 3]

It is electron micrograph of synthesized zeolite by a combination of two patents.

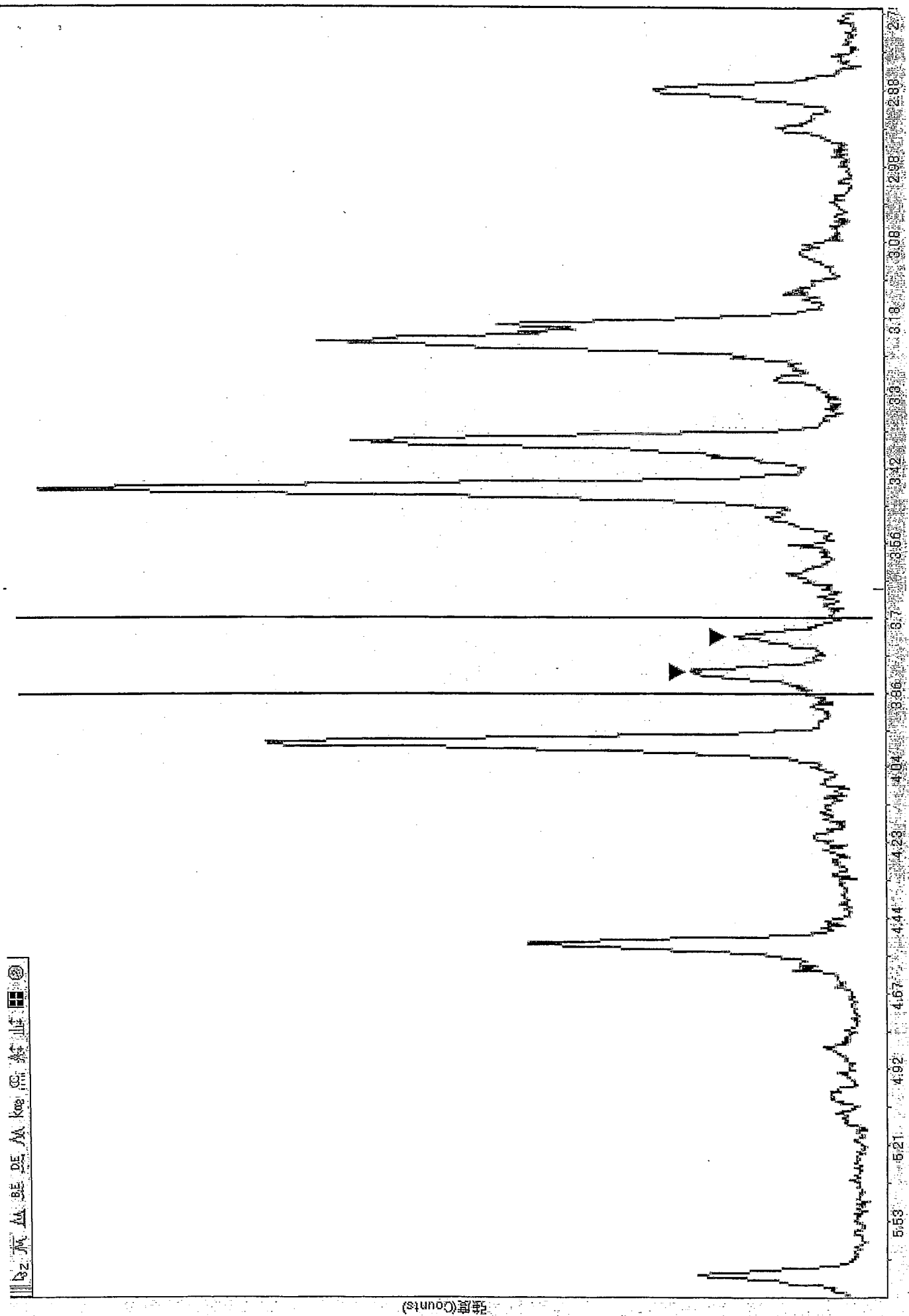






FIG.2

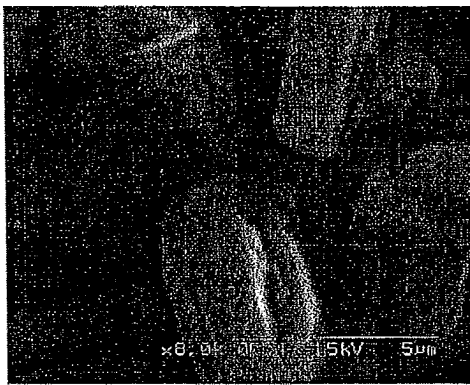


FIG.3